Army Research Laboratory

Predictive Capability for Hot Spot Ignition of Double Base Propellants

by Stephan R. Bilyk

ARL-RP-261 **August 2009**

A reprint from the *2006 Insensitive Munitions and Energetic Materials Technology Symposium*, Bristol, UK, 24–28 April 2006.

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14. ABSTRACT

Propellants are almost always ignited due to thermal processes. They can be ignited by direct application of heat or by the conversion of mechanical or electrical energy to heat. However, it is not necessary to heat the bulk energetic for ignition. Local regions which achieve high temperatures, so called “hot spots”, are sufficient to cause rapid decomposition and reaction. For “critical” hot spots, the reaction in the localized region must produce heat faster than the heat transferred to the material and losses to the surrounding environment. Otherwise, the hot spot cools and can eventually stop reacting. In their monograph work on the topic, Bowden and Yoffe (1952) estimated critical hot spots at the micron (0.1 to 10µm) length scale, with duration of 10^{-5} to 10^{-3}s and reaching 700K. The current research exercises a hydrocode to determine its ability to predict critical hot spot initiation of energetic materials resulting from thermo-mechanical coupling. For the simulations, the viscoSCRAM constitutive model was used to describe viscoelasticity, viscoplasticity, cracking and ignition in a double-base propellant when subjected to dynamic shear loading conditions. The effect of hot spot size and duration on the ignition threshold temperature was examined. The validity of the constitutive relations and the failure criterion are determined based on their ability to predict the observed mechanical response.

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Propellants are almost always ignited due to thermal processes. They can be ignited by direct application of heat or by the conversion of mechanical or electrical energy to heat. However, it is not necessary to heat the bulk energetic for ignition. Local regions which achieve high temperatures, so called "hot spots", are sufficient to cause rapid decomposition and reaction. For "critical" hot spots, the reaction in the localized region must produce heat faster than the heat transferred to the material and losses to the surrounding environment. Otherwise, the hot spot cools and can eventually stop reacting. In their monograph work on the topic, Bowden and Yoffe (1952) estimated critical hot spots at the micron (0.1 to 10μm) length scale, with duration of 10^{-5} to 10^{-3}s and reaching 700K. The current research exercises a hydrocode to determine its ability to predict critical hot spot initiation of energetic materials resulting from thermo-mechanical coupling. For the simulations, the viscoSCRAM constitutive model was used to describe viscoelasticity, viscoplasticity, cracking and ignition in a double-base propellant when subjected to dynamic shear loading conditions. The effect of hot spot size and duration on the ignition threshold temperature was examined. The validity of the constitutive relations and the failure criterion are determined based on their ability to predict the observed mechanical response.

INTRODUCTION

Energetic materials are often ranked in terms of their sensitivity when subjected to shock, shear, and thermal stimuli. The goal for military applications is to develop initiation criteria under each stimuli as well as a fundamental understanding of coupled behavior. Several useful analytical models and experiments already exist for shock and thermal stimuli. However, initiation due to shear loading is complex and poorly understood. Many hazardous scenarios such as hot metal fragments impacting an explosive canister can lead to shear initiation of an energetic. Shear initiation occurs at timescales over tens or hundreds of microseconds, an order of magnitude larger than shock loading. Energy is deposited in localized regions causing a local temperature rise, which for some energetics can even lead to the development of adiabatic shear bands.

It is generally accepted that initiation of an energetic is a thermal process [1]. High pressure accelerates chemical reactions, but most often does not

initiate them. Therefore, critical factors to initiate reaction are those that generate heat by direct application or by the conversion of mechanical or electrical energy to heat. This paper describes *non-shock mechanical stimuli* sufficient to create local regions, so called “hot spots”, which can lead to thermal ignition. By non-shock ignition we mean that there is an energy release but no shock wave. The energy release however, can approach that of a detonation.

For thermal ignition due to mechanical stimulus, it is not necessary to heat the bulk of the energetic since the locally created hot spots may reach sufficiently high temperatures. Energetic materials are a heterogeneous mixture of polycrystalline explosive, binder, and additives including voids created during material processing. Mechanical loading can nucleate hot spots (commonly in void regions) but only a few become critical hot spots. These critical hot spots ignite the energetic if the generation of heat in the localized volume is greater than the heat lost to the surroundings. In their monograph research on the topic, Bowden and Joffe [2] estimated critical hot spot parameters as typically of micron size (0.1-10μm), lasting for 10μs to 1ms, and reaching temperatures of approximately 700K. Clearly, if local temperatures are high, the size and duration can be smaller. Hot spots form during the interaction of stress waves with material defects and depend on the mechanical, thermal and chemical properties of the energetic. There are different mechanisms at the microstructural length scale that can create hot spot ignition. These include jetting of material grains, hydrodynamic pore collapse, viscous heating, shear localization, friction between grains, internal shear and shock interaction with second phase particles [3,4]. The dominant mechanism for producing the hot spot has not been generally agreed upon. However, Dienes [5] analytically showed that the largest contribution to potential heat generation is the frictional forces on shear crack surfaces.

Double base propellants are composed of nitrocellulose and stirred with a reactive plasticizer liquid nitrate ester such as nitroglycerine which also affects the oxygen balance. Stabilizers and gelatinizers are often added and the paste is hot rolled processed and pressed without the use of a solvent. The plasticizer is used to adjust the oxygen balance which affects the energy output and reaction temperature [6]. This class of propellant powders is often used in large caliber guns and solid rockets.

Initially, the activator punch test was developed to study shear initiation [7]. This test was limited since it was difficult to control the shear velocity independently of the pressure and the pressure on the shear surface was not well known. Recently, Krzewinski, *et al.* [8] developed a shear punch test at ARL. The shear punch test uses a modified Kolsky bar technique and obtains data for shear initiation of energetic materials subjected to dynamic loading conditions. In addition, some non-energetic polymer materials such as polycarbonate (PC) have been used as surrogate specimens for comparison purposes.

This paper first establishes an effective numerical modeling approach of the shear punch test. As an initial approach we chose to neglect the energetic properties of the material and focus on modeling the entire experiment with the severe deformations of the specimen. For this reason, the initial results

discussed are for a PC specimen. Next, we used a constitutive model that included a hot spot ignition criterion for PBX and a double base propellant.

EXPERIMENTAL DESCRIPTION

The apparatus used for the shear punch test was a modified Kolsky bar, as shown in Figure 1. The striker, incident, and output bars were 1.27cm diameter 350-maraging steel. The incident and output bars were 150cm in length, while the striker bar was available in 25, 50, and 55cm lengths. The varying striker lengths gave nominal pulse durations of 100, 200, and 220μs, respectively [8]. The specimen had a diameter of 1.905cm and a length of 1.27cm. As the compression wave travels during the entire test, the striker, input and output bars and the holder remain elastic. The specimen is the only material that undergoes plastic deformation.

The experimental measurements are also shown (boxed) in Figure 1. Impact velocity was measured using three fiber optic wires and an optical detector. Two strain gages were mounted near the center of the input and output bars to measure the incident, reflected, and transmitted strains. Finally, a scanning electron microscope (SEM) was used to measure the punch and dent displacements of the specimen as well as examine any fracture regions.

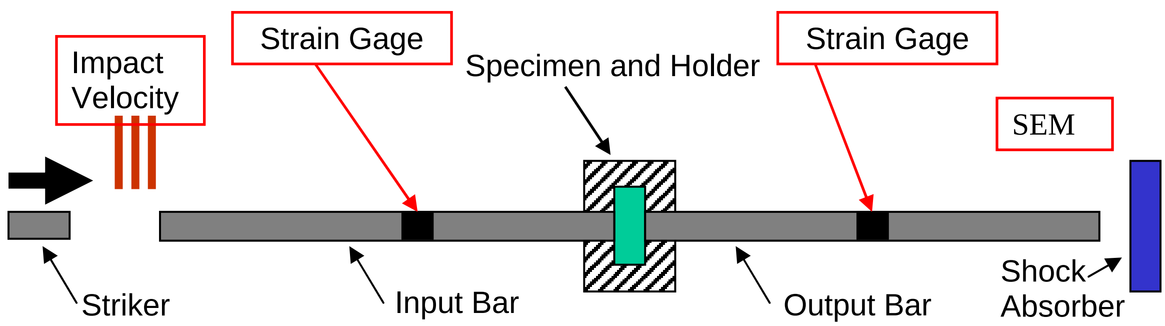

Figure 1. Schematic of Shear Punch Test and data collection (Not to Scale).

A special shock absorber and transfer piston (not shown in Figure 1) were designed to prevent reverse bar motion whenever the specimen reacted violently. Thin polyethylene disks were also placed between the specimen and incident/output bars for impedance matching. Copper (3mil) and Kaptan (5mil) disks were placed between the striker and incident bar to reduce ringing and wave shape a nearly rectangular incident compressive pulse. The specimen holder was made from 17-4 PH stainless steel and consisted of three pieces held together with six high-strength bolts. In addition, vacuum grease was applied between the specimen and specimen holder to fill any voids and reduce friction at the interfaces. With the applied grease, one can conclude that all initiations occurred because of the shearing within the specimen.

A typical deformed specimen shape is shown in Figure 2. The specimen shown is a double-base propellant, P1. Note also in Figure 2, that the shear surface has localized and runs along the outer radial edges of the incident bar.

For this dynamic test, the loading on the P1 specimen was great enough to eventually fracture the specimen along the shear surface.

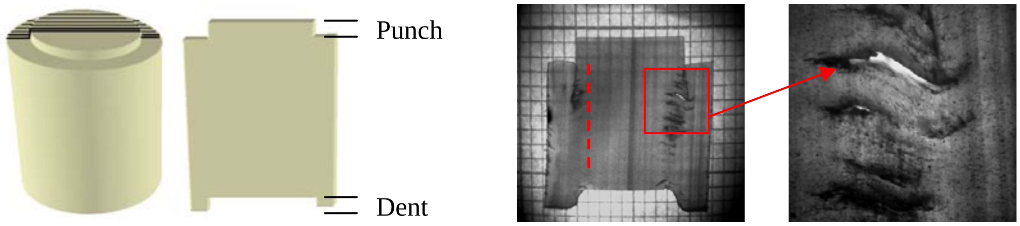

Figure 2. Typical specimen deformation and idealized shear surface (dotted line).

NUMERICAL DESCRIPTION

Numerical modeling of this test is difficult because it requires a mesh formulation that can withstand severe deformation and an ignition model that includes shear loading, damage, and frictional effects. In a pure Eulerian formulation, the material moves through a static mesh. Although a pure Eulerian formulation is not appropriate to study wave propagation, it is attractive because it can handle severe deformations. However, the material advection algorithm tends to "smear" the deformation over a number of cells leading to an unrealistic deformation. Further refinement of the mesh does not resolve material advection and creates an unreasonably large mesh for computational processing.

In a pure Lagrangian formulation, the mesh moves with the material. This formulation adequately describes the wave propagation but cannot handle the severe deformations of the specimen in a dynamic punch test. For this reason, an alternative formulation called an Arbitrary-Lagrangian-Eulerian (ALE) method was chosen. The ALE method starts out Lagrangian until severe deformations are triggered. At this time the Lagrangian formulation pauses to allow for some material advection and re-meshing, then returns to a Lagrangian formulation for the next time step. The advantage is that numerical dissipation is avoided until large deformations occur and then is limited to only those regions where there are severe mesh distortions and the mesh must be removed. The general name for the ALE method is adaptive mesh refinement since the mesh adapts to the materials' loading environment. The entire computational domain included the incident and output bars, the specimen, and the specimen holder. For the simulations presented, the 50cm striker bar was replaced with a prescribed input velocity boundary condition on the end nodes of the incident bar. The z-velocity pulse had a 1300m/s material velocity, a 5μs rise time with duration of 200μs.

A hybrid computational domain was also built for the simulations using 8-node hexagonal elements. Slide surfaces and symmetry conditions were also used to create the ¼-symmetry, butterfly computational domain, as shown in Figure 3. After a series of iterations, it was determined that the specimen and holder were best modeled with an Eulerian mesh. However, with new slide surface hydrocode capability, we feel that the specimen can be modeled with an ALE grid. The end portions of the input/output bars touching the specimen were assigned as ALE to transition from the Eulerian-only specimen to the Lagrangian-

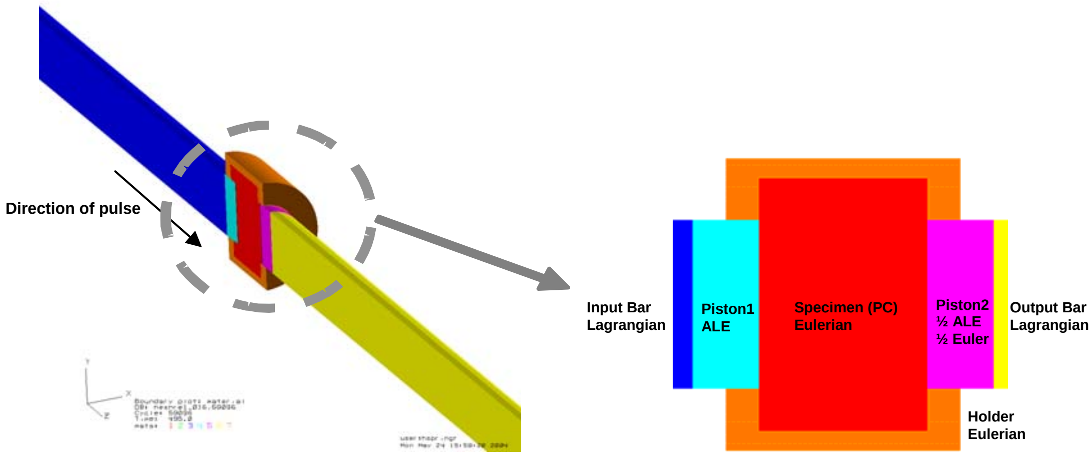

Figure 3. The hybrid computational domain for the shear punch test.

only input/output bars. The input bar, output bar and specimen holder were modeled using the previously mentioned elastic-plastic description. The specimen constitutive behavior was described using the viscoSCRAM model.

RESULTS AND DISCUSSION

A plot of the deformed mesh for the specimen is shown in Figure 4. Note the localized shear surface that formed in the specimen. The localized strain in the specimen emanated from the periphery of the indenting piece and, later in time, formed on the distal end at the holder/output bar interface. Figure 4 also shows a plot of the surrogate specimen temperature at the end of the simulation. PC has a melt temperature of 558 K. The temperature rise is due to the conversion of plastic work to heat. Although the temperature localizes near the bar/specimen interface, it dissipates to neighboring elements because of the mesh resolution. For a finer mesh, the temperature may localize along the idealized shear surface and reach a higher order of magnitude.

The specimen geometry is different from what is required in a conventional Kolsky bar. For this reason the strain rate is not uniform in the shear punch specimen. The specimen's strain rate reaches ~8000-9000s^{-1} and localizes along the idealized shear surface. An examination of the shear stress in the specimen during compressive loading at 600μs, shows the stresses reach 40-50 MPa. By comparison the principal compressive stress reaches ~150MPa in the center region and ~300MPa in the outer region. Of course, the state of stress in the specimen will change at the arrival of the transmitted wave. For a finer specimen mesh resolution subjected to this complex state of stress, the specimen material may form adiabatic shear bands. We also note that the pressure in the PC specimen reaches approximately -5MPa (tensile hydrostatic stress). This pressure is above the fracture pressure (-80MPa) therefore, the PC specimen did not fracture in this simulation.

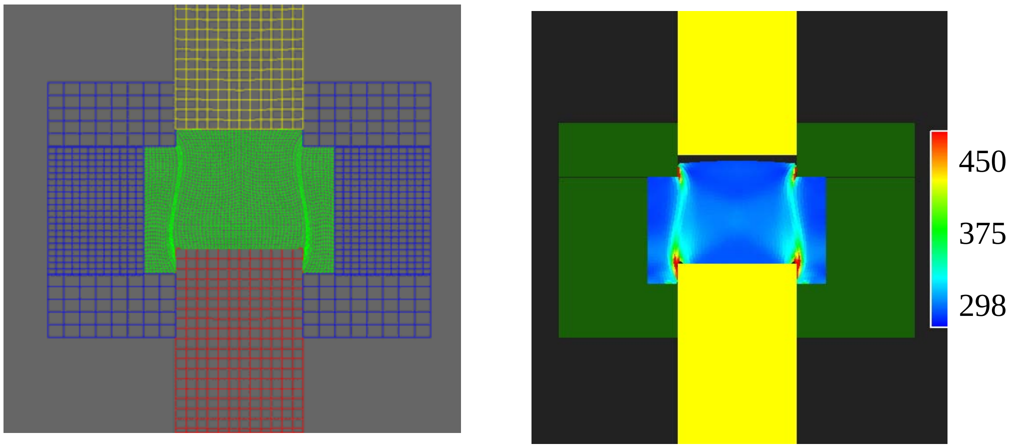

Figure 4. Final deformed shape and temperature [K] in the surrogate polycarbonate (PC) specimen.

A comparison of the strain gage signals to the observed result shows excellent agreement, as shown in Figures 5a and 5b. The incident and reflected pulses are shown in Figure 5a. The small difference in magnitude for the incident pulse occurs because the experimental impact velocity was 27.61m/s compared with 26.0m/s used in the simulation. The curvature at the beginning of the experimental input pulse is due to wave shapers added in front of the input bar. There were no wave shapers added in the numerical simulations. The ringing seen at the beginning and end of the numerical incident signal are due to the sharp discontinuity of the prescribed velocity boundary condition. Smoothing this boundary condition will reduce the ringing.

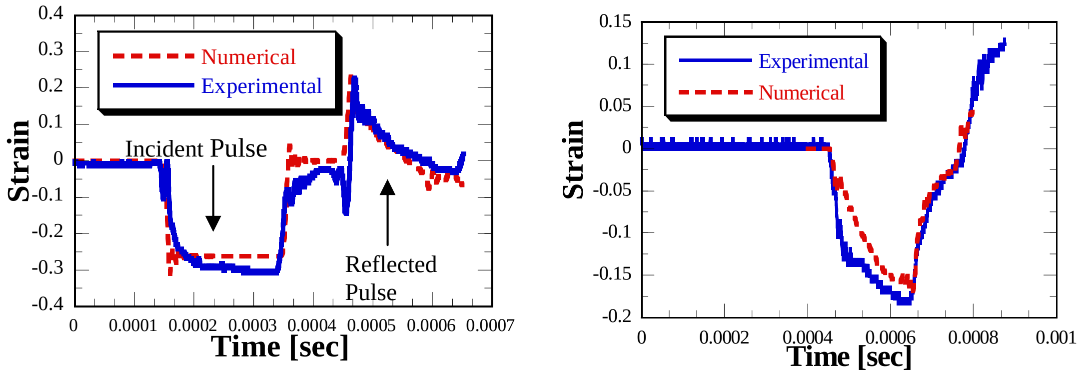

Figure 5. Comparison of strain signals for (a) incident bar strain gage and (b) the transmitted strain signal.

HOT SPOT INITIATION MODEL

For the simulations, the constitutive behavior of the specimen was modeled using viscoSCRAM [9]. The model captures rate dependence (linear-viscoelastic), damage accumulation (statistical-crack-mechanics), adiabatic mechanical heating and chemical heating that are apparent for some energetics.

Furthermore, the model does not include heat conduction because it is too slow compared to the deformation time scale.

The mechanical response has two constitutive assumptions. The first is that the strain rate can be decoupled into viscoelastic and deviatoric material damage components. The second is that the shear stress is determined from the viscoelastic strain rate. The viscoelastic portion is based on the work of Addessio and Johnson [10] and the damage model uses the statistical crack mechanics (SCRAM) approach of Dienes [11]. In addition to the aforementioned assumptions, the failure model assumes that for each element a micro-crack exists normal to the direction of the maximum (principal) deformation rate, as shown in Figure 6.

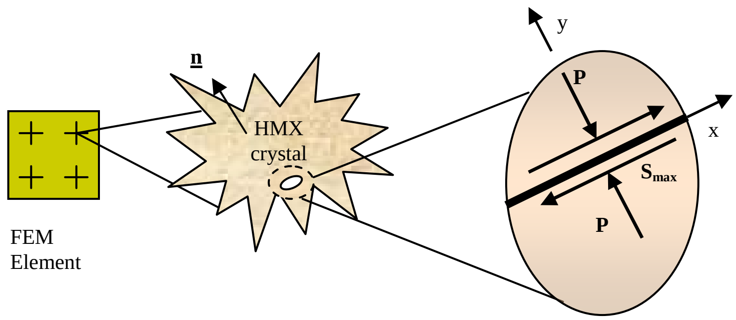

Figure 6. The viscoSCRAM hot spot model showing friction generated along a crack face.

Thermal heating in viscoSCRAM includes bulk heating at the continuum length scale and hot spot heating at the microstructural length scale. Bulk heating includes mechanical terms describing viscous, damage, and adiabatic volume change as well as, a chemical decomposition term. Chemical decomposition is based on Arrhenius first order chemical kinetics. For the continuum, the rate of temperature change with respect to time is written as

$$\dot{T} = -\gamma T \dot{\varepsilon}_{kk} + \frac{\Im}{\rho c_v}\left[\left(\dot{W}\right)_{ve} + \left(\dot{W}\right)_{cr}\right] + P_{he}\dot{q}_{ch} \tag{1}$$

where the first term on the right hand side represents adiabatic compression heating rate, the second term represents the inelastic work rates due to viscoelastic effects and cracking damage, and the third term is the bulk chemical heating rate.

The ignition criterion in the viscoSCRAM hot spot model describes frictional heating due to crack faces sliding. Given the stresses from two adjacent elements, the local strain energy release rate is determined. Then, at the end of a time step in the simulation, the change in crack length of the interface crack is determined. If the interface crack grows to be wider than the length of the element edge, the interface fails and is allowed to separate by not enforcing the constraints on the adjacent interface nodes. As the simulation progresses, the failed interfaces coalesce into macroscopic cracks. Once the shear stress exceeds a slip criterion, the adjacent crack faces are assumed to slip. The work

done by the slipping faces will generate heat and possibly ignite the energetic. This frictionally triggered hot spot model is included in the energy balance on a differential material volume near the crack face along with mechanical and chemical heating terms (neglecting heat conduction). Referring to Figure 6, the heat transfer near the 1-D crack face is given by

$$\rho_f c_f \dot{T} = k_f T_{,ii} + \rho_f \Delta H Z e^{-\frac{E}{RT}} + \mu_{static} p \dot{\varepsilon}_{ij} , \qquad l_f \geq y \geq 0 \qquad (2)$$

$$\rho_s c_s \dot{T} = k_s T_{,ii} + \rho_s \Delta H Z e^{-\frac{E}{RT}} , \qquad y > l_f \qquad (3)$$

where l_f is the hot spot length scale and μ_{static} is the coefficient of static friction. In eqns. (2) and (3), the left hand side is the heat stored in the region of the hot spot. The first term on the right side is the heat conducted away from the hot spot and the second term is the chemical heat generation per unit volume. For each finite element, the deviatoric stress is found on a plane normal to the direction of the maximum principal deformation rate. If the maximum shear stress exceeds the value of $\mu_{static} p$ then the crack is assumed to slip and generate heat. Note that p is the compressive pressure and if it is positive the crack is open and will not generate heat.

The viscoSCRAM constitutive model described was used to represent the behavior of the energetic specimen in the dynamic shear punch test, as shown in Figure 7. First, a plastic bonded explosive, PBX9501, was tried because of available material parameters. PBX-9501 is a brittle material and a very load sensitive energetic [12]. New crack faces are created during the early loading stages. As a result, Figure 7 illustrates that PBX-9501 generates heat due to chemical decomposition shortly after the arrival of the dynamic compression wave. For the double base propellant, the specimen experienced more plastic deformation and cracking (resembled an extrusion process) before it generated heat from chemical decomposition. Further work is required to validate the ductile materials parameters used for the double base propellant and reduce the numerical advection of the specimen. This will influence the generation of failure surfaces in the double base simulations to closer resemble the experimental photographs of Figure 2b which will in turn influence the shear ignition criteria.

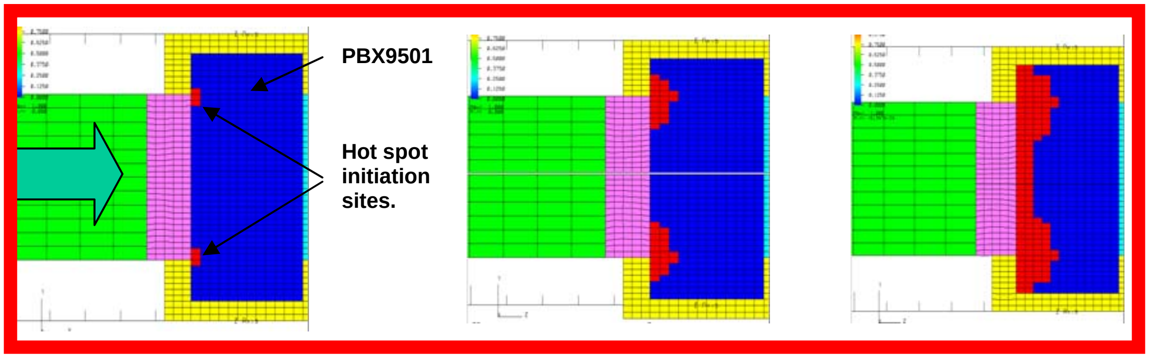

Figure 7. Chemical heat generation using the viscoSCRAM model.

SUMMARY AND OBSERVATIONS

A numerical model of a shear punch test has been developed to study the effects of shear loading on various energetics. To date we have completed simulations for nonenergetic polymer materials, plastic bonded explosives, and double base propellants. The simulations showed excellent agreement of the strain gage signals and showed the general trend of an idealized shear surface in the specimen. The hybrid mesh capability enabled complete modeling of the shear punch test. The Lagrangian formulation used for the incident bar and output bar provided an efficient solution to wave propagation. The ALE mesh for the specimen prevented hourglassing and excessive material advection while maintaining a reasonable timestep. More work is needed to reduce the advection in the specimen for the simulations, i.e. make the specimen more Lagrangian.

The prescribed velocity boundary condition eliminated the need to model the striker bar. Smoothing this boundary condition will reduce ringing in the incident strain signal. The simulations predicted the “punch” and “dent” material response in good agreement with observed results.

The hot spot shear initiation model was included in viscoSCRAM for PBX-9501. The simulation predicted chemical heat generation at the early stages during the arrival of the dynamic compression wave. It is emphasized however, that further work is required on determining the sensitivity of viscoSCRAM input parameters. Also, a clearer methodology used for developing material parameters for PBX-9501 is being completed for double base propellants. Furthermore, the author believes that with these additional material charaterizations and calibrations, the viscoSCRAM model will be a very useful tool for predicting insensitive munition behavior.

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